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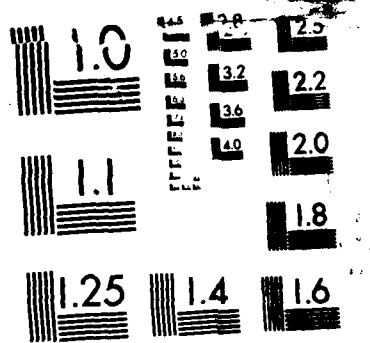
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ABSTRACT: A detailed study of the interaction of DNA polymer disolved in aqueous medium has been performed. This study takes accurate account of the distribution and dynamics of other disolved ions - counterions - as well as some aspects of the molecular organization of the nearby solvent. A theory of observed strong, sharp resonances in the microwave frequency range has been developed. Using numbers from independent experiments in the model developed gives a theory in full agreement with the measured resonant frequencies.

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I. Project History

We at the Purdue Physics Department have been involved in the theory of DNA as an interesting physical system for over 10 years. The special viewpoint which we have maintained has always been the power of the normal mode analysis of DNA polymer chain dynamics. This has proved to be a very fruitful approach to DNA physics.

We have available here an extensive library of software and data files for doing normal mode calculations in various DNA polymers. It is also easy and quick for us to update our database when new information becomes available.

This particular project began a year and a half ago. There were two complementary aspects to the proposal. First was a study of the effect of counterions and electrical activity of the solvent for dissolved DNA's. We anticipated that the absorption of electromagnetic radiation in the radio/microwave frequency region would be influenced by the counterion cloud surrounding the dissolved acid molecules. The second aspect of the proposal concerned the dynamics of localized irregularities along the polymer chain. We sought to develop and apply improved methods for studying the spectroscopy of localized modes. We planned to apply these new algorithms to determine the nature of the characteristic vibrations of localized defects, their interactions with the long chain normal modes, and, where appropriate, properties like their ability to grow or diffuse along the chain.

As this project was getting started, unusual new experimental results on microwave absorption of definite length DNA chain solutions were reported by the Maryland/EPA group. Specifically, Edwards, Swicord, Davis & Saffer[1] reported measurements of well defined resonances corresponding to "organ pipe" modes, longitudinal vibrations of the polymer molecules specifically corresponding in resonant frequencies to various integer multiples of vibrational half wavelengths on the chain as a unit. The characteristic lifetimes of these vibrational resonances as measured were much longer than anyone would have thought possible, judging by earlier work on the dynamical properties of DNA - solvent interactions. Dorfman and Van Zandt[2] had just recently published some papers estimating these very vibrational lifetimes, and had obtained shorter times by two orders of magnitude than those implied by the Edwards measurements. Emphasis was given in the execution of this project to delving into this contradiction.

Some months later, Alwyn Scott[3] and collaborators applied the nonlinear elasticity theory of "solitons" to this same experimental anomaly. They claimed considerable success, noting an extensive list of qualitative correspondences between the behavior of their "Boussinesq" solitons and the experimental observations. Some portions of our effort were devoted to a further study of the soliton hypothesis.

Theoretical Research on the Physical Properties of DNA

Lonnie L. Van Zandt
Department of Physics
Purdue University
W. Lafayette, Indiana 47904

April, 1986

ABSTRACT: A detailed study of the interaction of DNA polymer
disolved in aqueous medium has been performed. This study takes
accurate account of the distribution and dynamics of other disolved
ions - counterions - as well as some aspects of the molecular
organization of the nearby solvent. A theory of observed strong,
sharp resonances in the microwave frequency range has been developed.
Using numbers from independent experiments in the model developed
gives a theory in full agreement with the measured resonant
frequencies.

II Project Results to Date: The Edwards Experiment.

The following consider.tions on DNA-electrolyte dynamics become
of much less interest if the soliton theory of molecular vibrations
provide an adequate explanation of the Edwards resonances. The
soliton theory as laid out by A. Scott and coworkers comes down in
the end to a characteristic dimensionless parameter constructed of
three molecular system constants: a parameter giving the
nonlinearlity of the inter-atomic forces, a dispersion parameter
giving the dependence of the speed of longitudinal sound on
frequency, and an amplitude characteristic of soliton excitations.
Scott has noted the work of the Purdue group, suggesting
that our numbers might serve to evaluate the extent of soliton
character and indeed, such an application has been made.

The evaluation of the soliton "figure of merit" based on our DNA
sound wave dispersion curves and atomic interaction models yields a
number 3 orders of magnitude too small to explain the Edwards
results as a soliton excitation[4]. Indeed, the nonlinear component of
the dynamic interactions seems to be a very minor thing. We must note
that Scott does not accede in these conclusions at last
communication, and this result should still be held as controversial.

Two publications have been submitted concerning this matter. The
first was an application of our numbers to the Scott formalism.
Scott's differences with us on this matter concern the proper way to
take into account the thermal background of excitations over and
above the single photon absorption process. The second is cast in the
form of a theorem showing that with any physically allowable set of
assumptions about the thermal background, the soliton hypothesis
fails.

The electrical behavior of electrolytes is a strong function
of the concentration of the charged ion species present. This truism
presents particular analytic difficulties for the case of the
charged, dissolved DNA chain. Because of the balance between
electrostatic forces and diffusion in the near neighborhood of the
polymer, there is a strong concentration gradient of the electrolyte
ions. The characteristic length associated with this concentration
variation is about 5 to 10 Angstroms, (the so-called Manning
condensation radius). Incoming electromagnetic radiation passes
through this counterion layer before impinging on the molecule.
Scattered radiation passes through this layer again on its way back
out into the solution. A proper solution of the scattering/absorption
problem requires the solution of the electromagnetic field equations
in this region where the parameters are strong functions of radial
distance. Additionally, the net absorption of the radiation as seen
from the point of view of the solution as a single "stuff" contains a
large contribution from ion currents in the solvent, energy which
never reaches the molecule.

A further complication is that the ions interact strongly with
the solvent, passing through it only with great difficulty;
inversely, any motion of the liquid drags the ions along, and is thus

associated with a considerable current density. The net result is that the electromagnetic field problem and the liquid dynamical problem and the ion dynamical problem are all inseparably linked. Note further that these systems are all described by vector fields. Reduced to coupled scalar differential equations, there are seven second order differential equations to be solved, the solution of each providing an inhomogeneous term for the next in the chain. Ancillary functions in the problem are obtained from the solution of the Poisson/Boltzmann relations for the ion densities as a function of radial distance.

This nest of interconnecting fields is connected to the dynamics of DNA in two ways: 1) the longitudinal component of the electric field arriving at the inner core of the system directly acts on the charges of the acid, and 2) the charged liquid at the interface between molecule and solvent acts mechanically on the polymer as the solvent is dragged back and forth by the fields. The first interaction is written down directly; the second proves to be best expressed as a boundary condition. The particular parameter proving most convenient for the expression of conditions at the interface was found to be the ratio of the incoming and the scattered electromagnetic field amplitudes.

The connection between the near dynamical system of liquid motions, electric fields, and molecule motion and the actual externally applied microwave field is made by a far - but not infinitely far - boundary condition on the incoming component of the electromagnetic problem.

Our analysis is much more complete and thereby more directly applicable than the earlier treatment of Dorfman and Van Zandt that neglected the solute ions. There are now fewer "dark corners" in which an anomaly might hide. Further, Dorfman and Van Zandt made the classical "wetting" assumption, that the motion of the liquid and the molecule are identical at the interface where they join.

When we too made the assumption of strict wetting of sticking boundary conditions at the interface, we similarly obtained a heavily damped description of the molecular motion. This damped solution is characterized by a low amplitude of molecular motion, no sign of any resonant behavior, and a low amplitude of electric field at the molecule. This last result is in accord with some other earlier studies in which the system was analyzed in terms of inhomogeneous dielectric theory[5]. Electromagnetically speaking, because of the impedance mismatch at the molecular surface, the DNA molecular space looks like little cells of vacuum to the irradiating applied field.

We examined the case of relaxing the strict sticking boundary condition. We assumed that accompanying the gross tendency of the molecule to follow the liquid, it was capable of an additional motion as if free. The linear nature of most of the descriptive equations allowed this to be done fairly simply. A small slipping component of the motion riding along on top of the sticking portion offers the possibility of explaining one Edwards experiment. Such a slipping component was added ad hoc to the theory. With this addendum in the theory, we were able to produce resonances like those observed.

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The extra degree of freedom involved in the slipping hypothesis arises out of the structured nature of the water in the polymer vicinity. When we make a division of the water motions into shells of nearest, next nearest, and progressively more distant neighbors shading ultimately into amorphous liquid at large distances, we introduce additional explicit degrees of freedom into the problem. These extra degrees of freedom are subsumed into the continuum field motion of the liquid right up to the polymer interface in the Dorfman/VanZandt model.

Breaking these extra degrees of freedom out of the water continuum would not produce any significant differences were it not for the fact that the nature of the water/water interactions and the water/polymer interactions are substantially different. The characteristic lifetime for thermally disrupting and reforming bonds on the molecular level differs by an order of magnitude for these two processes. Hence, the last layer of water right up against the DNA behaves much differently from the others. The combination of DNA and last water layer as a dynamical system has a long lived, weakly damped degree of freedom in addition to a conventionally damped one. The application of reasonable numbers taken from independent experiments to this system produces a striking fit to Edwards' most recent data. This is more fully described in our most recently submitted communication.

III The Local Mode Problem.

In 1979, Putnam and Van Zandt[6] published a determination of some portions of the absorption spectrum of a terminus to a DNA polymer chain. Vibrations peculiar to the end of a chain and distinct from those of the bulk, far from an end, are a degenerate case of what is known in crystal physics as surface waves[7]. The unit cell in our case is a full monomer unit, structurally much more complex than a simple crystal unit cell, but the two dimensional k space descriptive of the crystal surface has shrunken to a single point; the relative structural richness of our complex unit cell compensates for the poverty of our transverse k space.

The method of Putnam and Van Zandt is costly in terms of computing time for the meager amount of information derived from its application. It is a "brute force" computation in which an expansion of the end behavior is carried out in terms of the infinite chain, "bulk" oscillation modes. A very fine grained description of the infinite chain modes is required. The precise details of why this should be so are not very interesting. Let it suffice to point out that the process is analogous to performing a Fourier transform by constructing one at a time the functions for an increasingly fine grained Fourier series.

At about the time this project was beginning, we realized that much of this difficulty might be avoided because the shape of the functions being constructed by the transform process could be determined analytically and the problem conceptually reduced to obtaining a few parameters for a function whose basic form was given. This phase of the project has not progressed to manuscript form as

yet, although the formal problems have been solved. The computer codes are in draft form.

Lindsay[8] has actually seen an end mode in Brillouin scattering at the theoretically predicted frequency, somewhat below 1 gigaHertz, but this news is stale by now, and no further experimental explorations have been done to show end modes in other regions of the spectrum. The experiments are difficult, and no further suggestions have been forthcoming from the theory to encourage them because of the tedium of executing the Putnam Van Zandt algorithms.

Work on this problem has progressed under this contract, but no publishable results have as yet been obtained.

References

(1) G.S. Edwards, C.C. Davis, J.D. Saffer, & M.L. Swicord, Phys. Rev. Lett. 53, 1284 (1984).

(2) B.H. Dorfman & L.L. Van Zandt, Biopolymers 20 833 (1983).

(3) A.C. Scott & J.H. Jensen, Phys. Lett. 109A 243 (1985).

(4) L.L. Van Zandt, Existence of Soliton Waves on DNA Polymer, submitted for publication.

(5) K. Foster, M.A. Stuchley, A. Kraszewski, S.S. Stuchley, Biopolymers 23 593 (1984).

(6) B.F. Putnam, L.L. Van Zandt, E.W. Prohofsky & W.N. Mei, Biophys. Jour. 35 271 (1981)

(7) R.F. Wallis, Int. Conf. on Local Excit. in Solids, N.Y. Plenum Press (1968).

(8) M.B. Hakim, S.M. Lindsay & J. Powell, Biopolymers 23 1185 (1984).

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